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doi:10.1289/ehp.7141 (available at http://dx.doi.org/)
Online 24 March 2005



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Acknowledgements: Funding for this project and manuscript preparation was provided in part by the National Center for Environmental Health (CDC), the American Water Works Asso. Research Foundation, and NIH-Nantional Cancer Institute-Occupational and Environmental Epidemiology Branch. No conflict of interest was reported.

We thank Charles Wilkes for his assistance in the design and implementation of the field study; Marielle Brinkman, Michael Holdren, William Keigley of Battelle for air and breath sample analysis; Russell Dietz of BNL for assisting in tracer analysis; Ben Blount, Michael Bonin, Lalith Silva, Mitchell Smith and Charles Dodson of CDC for assisting in blood analysis; Erika DePaz of UNC-Chapel Hill for water analysis support; and our nurses and field data collection staff for their hard work.

Running Title: Household Water Use Activities and Trihalomethane Exposure

Key Words: trihalomethane, chlorination, water use, disinfection byproducts, exposure,

biomarkers

List of Abbreviations:

American Water Works Association Research Foundation AwwaRF National Center for Environmental Health **NCEH** Centers for Disease Control and Prevention CDC National Cancer Institute NCI trihalomethane THM total trihalomethane TTHM chloroform CHCl₃ bromodichloromethane CHBrCl₂ dibromochloromethane CHBr₂Cl bromoform CHBr₃ **Environmental Protection Agency USEPA** North Carolina NC **Texas** TXBrookhaven National Laboratory BNL gas chromatography/mass spectrometry GC/MS single breath canister SBC glutathione S-transferase -theta-1 GSTT1 confidence limits CL neural tube defects **NTDs**

Outline of manuscript section headers

I. Abstract II. Introduction III. Methods a. Study location/ participants b. Data collection c. Water samples d. Air samples e. Blood samples Breath samples g. Data analysis IV. Results a. Water supply temperature and THM concentration b. Ambient air temperature and THM concentration c. Markers of exposure – blood and exhaled air THM V. Discussion VI. References **Tables** Figures Figure Titles and Legends

Abstract

Individual exposure to trihalomethanes (THM) in tap water can occur through ingestion, inhalation or dermal exposure. Studies indicate that activities associated with inhaled or dermal exposure routes result in a greater increase in blood THM concentration than ingestion. We measured blood and exhaled air concentrations of THM as biomarkers of exposure to participants conducting 14 common household water use activities, including ingestion of hot and cold tap water beverages, showering, clothes washing, hand washing, bathing, dish washing, and indirect shower exposure. We conducted our study at a single residence in each of two water utility service areas, one with relatively high and the other low total THM in the residence tap water. To maintain a consistent exposure environment for 7 participants, we controlled water use activities, exposure time, air exchange, water flow and temperature, and non-study THM sources to the indoor air. We collected reference samples for water supply and air (pre-water use activity), as well as tap water, and ambient air samples. We collected blood samples before and after each activity and exhaled breath samples, baseline and post-activity. All hot water use activities yielded a 2-fold increase in blood or breath THM concentrations for at least one individual. The greatest observed increase in blood and exhaled breath THM concentration in any participant was due to showering (direct and indirect), bathing, and hand dishwashing. Average increase in blood THM concentration ranged from 57 to 358 pg/mL due to these activities. More research is needed to determine whether acute and frequent exposures to THM at these concentrations have public health implications. Further research is also needed in designing epidemiologic studies that minimize data collection burden, yet maximize accuracy in classification of dermal and inhalation THM exposure during hot water use activities.

Introduction

Trihalomethanes (THM) are a byproduct of water chlorination, arising from the reaction between natural organic matter in the source water and chlorine used for disinfection. There are four primary species of THM: Chloroform (CHCl₃), bromodichloromethane (CHBrCl₂), dibromochloromethane (CHBr₂Cl), and bromoform (CHBr₃). The speciation of the THM depends on raw water quality and treatment characteristics (Miles et al. 2002). The US Environmental Protection Agency has established a maximum contaminant level of 0.08 mg/L for the total THM due to increased evidence of adverse health effects linked to these compounds (USEPA 1998). Researchers have found an association between elevated levels of THM and adverse health outcomes, including cancer (Cantor et al. 1978, 1987, 1998; Hildesheim et al. 1997; King and Marrett 1996; McGeehin et al. 1993) and adverse reproductive outcomes (Aschengrau et al. 1989, 1993; Bove et al. 1995; Gallagher et al. 1998; Klotz and Pyrch 1999; Waller et al. 1998). Exposure assessments for most of these studies were based on reported levels of total THM in the water distribution system serving the participants' residences, and in some cases reconstructing study participants' water consumption histories.

Exposure to THM through routes other than ingestion has been demonstrated as significant components of the overall exposure matrix. In controlled experiments, Weisel et al. (1992) and Xu and Weisel (2005) reported elevated breath concentrations of CHCl₃ due to showering. In a later field study of 33 subjects using public water supplies in New Jersey with relatively low THM concentrations, Weisel et al. (1999) determined that timing of sampling post-shower exhaled air was important in order to capture a high correlation to water concentration. Critical

time frames reported by their study were 20 minutes for CHCl₃ and CHBrCl₂, and 5 minutes for CHBr₂Cl and CHBr₃.

Weisel and Jo (1996) demonstrated that dermal contact is an important route of exposure for CHCl₃, reporting higher exhaled air concentrations from this route than from inhalation due to showering and bathing. Gordon et al. (1998) also reported elevated CHCl₃ concentrations in exhaled breath from subjects that breathed clean air while bathing in waters ranging in temperature from 30 to 40°C (86 - 104°F). For these dermal-only exposures, they reported that for similar levels of CHCl₃ in the bath water, much higher levels of the compound in exhaled air were measured from an individual taking a 40°C bath as compared to the same individual taking a 30 or 35°C bath.

Studies have demonstrated that exposure to THM results in significant increases in blood THM concentrations. Backer et al. (2000) reported increases in blood CHCl₃, CHBrCl₂, and CHBr₂Cl compared to pre-activity blood levels in groups of approximately 10 individuals each due to showering, bathing, and consuming 1 liter of cold tap water for a 10-minute period. They found the increases in blood concentrations of these THM from showering or bathing were significantly greater than the increases from drinking 1 liter of water. Pegram et al. (2002) reported maximum blood concentrations of CHBrCl₂ ranging from 0.4 – 4 ng/mL due to ingestion versus 39 - 170 ng/mL due to dermal contact with water containing the same concentration of CHBrCl₂. They also reported that blood CHBrCl₂ levels returned much more rapidly to baseline after ingestion (4 hours) as opposed to dermal exposure (24 hours). Lynberg et al. (2001) measured THM in pre- and post-shower blood samples from 25 participants in each of two water utility service areas. They reported significant inter-site differences in both tap

water samples and blood THM levels, as well as significant increases in blood THM levels for all participants due to the showering event. Miles et al. (2002) further analyzed the data from the field study, and found that while showering activity shifted the THM distribution in the blood towards that found in the corresponding tap water (including concentration), there was no significant correlation between blood concentration and tap water concentration.

Household water uses other than showering and bathing have not been evaluated in terms of potential exposure to THM. In this study, we determine the relative contributions of showering and bathing, along with twelve other water use activities, to THM exposure in a household environment. The purpose of this paper is to provide a description of the methods used in our study, and a summary of the results. The findings are relevant to the design and implementation of epidemiological studies concerning exposure to volatile water supply contaminants.

Methods

Study location/ participants: We conducted our study at a single residence in each of two sites: one in North Carolina (NC Site), and the other in Texas (TX Site). The floor plans for the study residences at the NC Site and TX Site were almost identical. Both were 3 bedroom/2 bathroom, one-story, ranch-style houses (about 111.5 m² or 1,200 ft² total floor space). The heating and ventilation systems in both residences were central air. Both had electric hot water heaters. Each residence was served by a public water distribution system. The study was conducted August 5 – September 17, 2002 in NC, and October 13 – November 6, 2002 in TX. We treated the data as representative of a water supply with relatively high (NC) and relatively low (TX) THM concentrations, predominated by chlorinated THM species.

We planned for and recruited seven participants by advertising in local media and distributing study flyers on local college campuses. We used a standardized questionnaire to screen applicants for the following eligibility criteria (acceptable range given in parentheses): age (18-35), body mass index (BMI; 22-24), tobacco smoking (non-smoker only), alcohol consumption (average < 2 drinks per day), swimming activity (< 4 days per week). We also excluded applicants who reported asthma or other breathing problems, high blood pressure or hypertension, a history of problems associated with blood draws, regularly taking any medications for any health conditions, or any condition that would prevent them from conducting the water use activities prescribed by our study. The final study group was composed of 3 males and 1 female at the NC site, and 1 male and 2 females at the TX site. The age range for participants in our study was 21-30. Two of the male participants at the NC site reported their race as African-American. All other participants reported their race as Caucasian.

Data collection: Prior to the introduction of participants, we prepared the study residence for data collection and analysis. Only one of the bathrooms in each residence was used as the study bathroom. Approximately 30 minutes before the 1st activity began each day, the second bathroom door was shut and the vent fan turned on. To prevent and account for contribution of THMs to household air, the use of the second bathroom during the study activities was minimized as much as possible and was documented. The showerhead in the study bathroom of each residence was replaced with a custom showerhead designed to maintain consistent flow. This showerhead was connected to a remote water sampling apparatus designed to minimize loss of volatile THM. The apparatus was used to collect water samples from the showerhead and the shower stall drain. The thermostat for central air conditioning (HVAC) in each house was set at 75°F, and the HVAC fan was set to the "on" position during the entire study period. The exhaust

fan in the study bathroom was not turned on at anytime during the study. At each study site, we conducted airflow and tracer gas studies to characterize the house-to-environment air exchange rates and bathroom-to-house air flow rates, and to identify the optimal locations for collecting household air samples during the THM exposure study (Dietz and Cote 1982).

We collected data THM exposure data over a two-day period for each study participant. The second day of the study typically occurred approximately one week after the first. On each day, the participant performed a set of prescribed water-use activities while we collected pre- and post-activity samples of air, water, blood, and exhaled breath. These activities are listed in Table 1. Between events on the participation day, the participant was required to remain in the residence. We designed the sampling regimen so that activities expected to result in the largest increase in internal dose levels were spaced at estimated time intervals sufficient to allow blood THM concentrations to return as much as possible to pre-exposure levels before the next water use activity. For some activities we collected concurrent air and/or water samples, as well as exhaled breath samples. Water temperature was measured during each activity.

To reduce the likelihood of inadvertent THM exposure, each participant arrived at the study residence the night before his/her scheduled day of data collection, and slept in the study residence. Upon arrival, the participant completed a questionnaire to provide information on demographics, water use and consumption in the past 48 hours, and exposure to chemicals that might be confounding factors in the study. These data were collected primarily to screen for water or chlorinated compound use (like swimming, etc) that could interfere with our premise that early morning blood concentrations could represent a "baseline" for each individual. The

subjects were instructed to wear swimsuits for the showering and bathing components of the study.

Over the study period, we measured the flow of water to each study house using a water meter data logger (Meter Master Model 100EL, F.S. Brainard Company, Burlington, NJ). These data were collected primarily for modeling purposes, and will be discussed in a separate manuscript. We measured ambient and indoor temperatures, and relative humidity using electronic thermometers. We controlled and standardized the water temperature for each study activity.

Water samples: We collected 21 water samples over the 2-day period. These samples were either associated with a water use activity or collected from a cold-water tap over the course of each exposure day to establish "baseline" THM concentrations (total and each of four species). We collected and analyzed duplicates of each sample. All water samples were collected using headspace-free 40-mL acid-washed glass vials. Immediately after collection, ammonium sulfate was added to the sample in order to quench residual chlorine and prevent further THM formation. We measured and recorded the temperature of the tap water for each sample. Sample containers were refrigerated and packed into coolers with ice packs and shipped by overnight express courier to the University of North Carolina at Chapel Hill for analysis using gas chromatography.

Air samples: We collected air samples to determine the levels of THM (total and each of the four species) in the air associated with each activity. Thirteen samples were collected over the two-day study period for each participant. We collected a "baseline" sample each day prior to any water use activity. The air samples were collected using pre-cleaned and evacuated SUMMA®-

polished 6-liter stainless steel canisters (Scientific Instrumentation Specialists, Moscow, Idaho, and Biospherics, Hillsboro, Oregon). We collected "grab" samples by opening the canister valve and allowing air to flow into the canister until atmospheric pressure equilibrium was attained (≤ 1 minute). We shipped exposed canisters by overnight express courier to Battelle Memorial Institute in Columbus, Ohio for analysis. Samples were analyzed by automated gas chromatography/mass spectrometry (GC/MS) using a modified version of U.S. EPA Method TO-14 (Winberry et al. 1990).

Blood samples: We collected blood samples from each participant in order to examine the levels of THM (total and each of four species) associated with each water use activity. Vacutainers (Becton, Dickinson & Co., Franklin Lakes, NJ) were prepared by heating, restoration of vacuum, and resterilization in order to eliminate background contamination from the blood collection device (Cardinali et al. 1995). We collected samples approximately 5 minutes before and after each activity, using a multi-sample adapter (venous catheter). Additional blood samples were collected 30 minutes following the shower and bath activities. The catheter remained in the participant for the duration of each day of the study, approximately 12 hrs. We collected a total of 26 10-ml blood samples from each participant over the course of the 2-day study, 14 on Day 1 and 12 on Day 2. After collection, each blood sample was refrigerated and packed into coolers with ice packs, and at the end of each day shipped by overnight express courier to the Volatile Organics Laboratory at the U.S. Centers for Disease Control and Prevention, in Atlanta, Georgia. We analyzed THM in the blood samples using a variation of the standardized method reported by Ashley et al. (1992). This method includes spiking 3-mL blood samples with isotopicallylabeled standards, extracting with solid-phase microextraction, and analysis by gas chromatography followed by high-resolution magnetic-sector mass spectrometry. We quantified blood THM concentrations using calibration curves generated from dilutions of pure samples of each THM species. Blanks and quality control materials were analyzed with each analytical run. Detection limits were in the parts-per-quadrillion range, allowing the quantification of most samples even at background levels.

Breath samples: We collected breath samples using a self-administered procedure in which the subject exhales alveolar air directly into an evacuated single breath canister (Pleil and Lindstrom 1995). For this study we used 1-L Silcosteel stainless steel canisters (Entech, Simi Valley, CA) fitted with a short Teflon tube that serves as a disposable mouthpiece. We instructed the subject to begin sample collection near the end of a normal resting tidal breath in order to provide what is mostly alveolar breath. We collected a total of 15 breath samples from each subject over the two-day study period. Baseline measurements were obtained once per day before all activities began. Samples were shipped at the end of each day by overnight express courier to Battelle for THM analysis (total and each of four species), which was carried out by the same automated GC/MS procedure used for air samples.

Data analysis: We calculated summary statistics (mean, standard deviation, median, range) for measured THM species in water, air, blood and exhaled breath samples, and for measurements of temperature in the water samples and in the ambient air during activities. We calculated relative exposure, defined as the ratio between pre- and post-activity blood concentration and between exhaled breath concentrations, for each participant and activity. We plotted the data, and examined for natural breakpoints. Based on this procedure, we established a cutpoint of 2-fold deviation from baseline concentrations as indicators of meaningful increase or decrease in these biological marker concentrations. We established similar criteria of \pm 20% for the ratio of

activity-related water concentrations to baseline (cold tap) water sample concentrations, and a 5-fold deviation in the ratio of activity-related air concentrations to baseline. Our approach is similar to that suggested by the American Chemical Society Committee on Environmental Improvement (ACSCEI) to determine whether increases in biological concentrations are meaningful when comparing environmental chemistry data (ACSCEI, 1980). ACSCEI suggested an increase of at least 3 times the standard deviation of the smallest (baseline) concentration in making this determination. Our approach is generally more conservative.

We used a repeated measures design of the general linear model (Ott and Longnecker 2001) to test for statistically significant inter-site, inter-participant, and temporal differences in measured water temperature and concentrations of THM in water. We used two-factor experiments with repeated measures on one factor (order of activity or baseline measurements as a proxy for time), and $\alpha = 0.05$ level of significance, to conduct these analyses.

Results

Water supply temperature and THM concentration: Figure 1 provides a summary of the median and range of concentrations of THM measured in baseline (cold tap) and water samples from each water use activity that resulted in at least a 2-fold increase in biological markers of exposure for at least one participant. It also includes the median water temperature for each sample type. The only activity that did not meet the criteria for inclusion in Figure 1 was ingestion of a cold tap water beverage on Day 1.

Baseline THM concentrations in the tap water were much higher at the NC site for total THM, ranging from 113 to 212 μ g/L compared to a range of 12 to 53 μ g/L at the TX site. Though these

concentrations did change over the course of the day, the difference between concentration for any THM by time of day was not statistically significant across the study population (p = 0.07 to 0.65). THM concentrations in the activity-associated water were also much higher at the NC site compared to the TX site.

Most ratios of THM concentration in activity-associated water to concentration in baseline (cold tap water) samples were near or below 1.0. Only the ratio for CHCl₃ for the showering event at the NC site exceeded our criteria of a 20% increase as being meaningful. At the NC site, median ratios of activity to baseline concentration for several THM species and activities were at least 20% less than 1.0, including CHBrCl₂ in showering and bathing, and CHBr₂Cl in showering and hand dishwashing. At the TX site, we did not observe a deviation of greater than 20% in ratios of THM concentration in activity and baseline water samples at the group or individual level, except for one participant where water for the shower, and for hand dishwashing had a ratio of 3.2 CHBr₂Cl and 3.3 TTHM, respectively.

We found that activity-associated water temperatures for most activities in Figure 1 were much higher than the temperature of the corresponding baseline water sample, with the exception of the automatic clothes washing activity. Median temperatures of the baseline (cold tap) water samples were very similar, with a difference of less than 2° C for any activity between the two study sites. The inter-site differences in the median water temperature were less than 1° C for most activities. We found no statistically significant correlation between water temperature and THM concentration, with the exception of CHBrCl₂ at the NC site (p = 0.02).

Air temperature and THM concentration: Table 2 provides a summary of median and range of concentrations of THM measured in baseline samples (prior to any water use activities) and in ambient air samples for each water use activity that resulted in at least a 2-fold increase in biological markers of exposure for at least one participant. It also includes the median and range in air temperature for each sample type. The only activity that did not meet the criteria for inclusion in Table 2 was ingestion of a cold tap water beverage on Day 1.

At both study sites, we observed a greater than 5-fold increase in the ratio of activity ambient air to baseline THM concentration for all THM compounds other than CHBr₃ for participants as a group due to showering and indirect shower exposure, and due to the bathing activity (except CHBr₂Cl). The air TTHM concentration during showering increased by 70% across individuals at the NC site, and by 38% at the TX site (data not shown). We observed a 4- to 11-fold (median = 7) increase in ambient air TTHM concentration due to the hand washing activity across participants at the NC site. This increase was primarily due to a corresponding increase in CHCl₃ concentration. We also observed large increases in ambient air CHCl₃ due to the automatic clothes washing with bleach (median increase > 9-fold) and the hand dishwashing (median > 5-fold) activities across participants at the TX site. For most of the other water use activities listed in Table 2, we observed a slight to moderate increase in ambient air THM concentration at both sites (median increase < 2.5-fold).

For the activities listed in Table 2, median temperatures of the baseline ambient air samples were equal for Day 1 and within 0.7°C for Day 2. Median temperatures of ambient air during the water use activities were within 5% of baseline at both sites, except for the clothes washing II

activity at the TX site. For that activity, the median air temperature was 27°C (81°F) compared to a median baseline temperature of 23°C (73°F).

Markers of exposure – blood and exhaled air THM: Table 3 provides a summary of median and range of concentrations of THM measured in blood samples collected 5 minutes before and after each water-related activity by study site. At both sites, there was a greater than 2-fold increase in blood concentrations for all participants and all THM species except CHBr₃ due to the showering and bathing activities. Increases as a result of showering were 5- to 15-fold in participants at the NC site and approximately 5-fold at the TX site. Increases as a result of the bathing activity were 3- to 6-fold in participants at the NC site, and 3- to 19-fold at the TX site. Hand dishwashing resulted in a 2- to 8-fold increase in blood THM concentrations (except CHBr₃) in 2 of the 3 participants at the Texas site. Increases of 3-fold in concentrations of CHBrCl₂ and CHCl₂Br were observed in the other participant. Hand dishwashing resulted in a less than 2-fold increase in blood THM concentrations in 3 of the 4 participants at the NC site.

The average pre-shower blood TTHM concentration at the NC and TX site were 47 and 19 pg/mL, respectively. The average increases in blood TTHM due to showering at the sites were 358 and 79 pg/mL, respectively. We observed similar pre-activity average blood TTHM concentrations for bathing, and hand dishwashing (except one participant at the TX site). The average increases in concentration for bathing were 164 and 118 pg/mL at the NC and TX sites, respectively. The average increases in concentration for hand dishwashing were 98 and 57 pg/mL, respectively, but there was a high degree of inter-participant variation at both sites. Increases in blood THM for the other activities were generally less than 20 pg/mL, and highly varied.

Table 4 provides a summary of the median and range of concentrations of THM in exhaled breath samples collected prior to all water use activities (baseline) and during or after activities by study site. The baseline exhaled breath THM concentrations were very similar between the two sites for all THM species except CHCl₃, which was consistently higher at the NC site. Baseline CHBrCl₂ concentration in one NC participant was 9 μg/m³, but this was inconsistent with all other baseline measurements at NC, which ranged from below detection limit (0.8) to 4.6 μg/m³.

We found a greater than 2-fold increase in the median exhaled breath concentrations of total THM across participants as a group due to bathing (both study sites) and showering (NC) activities, and an almost 2-fold increase due to showering at the TX site. These increases in TTHM were primarily due to increases in CHCl₃ concentration. Similar increases in median exhaled breath concentrations of CHCl₃ were also observed due to hand dishwashing activities at both sites, the automatic dishwashing activity at the TX site, and the automatic clothes washing with bleach activity at the NC site. Across individual participants, increases in exhaled breath TTHM concentrations due to showering ranged from 3 to 6-fold at the NC site, and were approximately 2-fold at the TX site. Individual increases due to bathing ranged from 3 to 6-fold at the NC site and 3 to 19-fold at the Texas site. Individual increases due to hand dishwashing ranged from approximately 1.5- to 2.5-fold at both sites, except for one outlier at the NC site with a measured decrease of 0.5-fold. This outlier had no influence on any of the reported results. We observed a 2-fold or better increase in the exhaled breath concentration of at least one THM compound in at least one study participant due to each of the other water use activities, with the exception of hand washing and indirect shower exposure.

Discussion

We measured blood and exhaled air concentrations of THM as biomarkers of exposure to participants conducting 14 common household water use activities (Table 1). We found that the showering (10 minutes) and bathing (20 minutes) activities consistently resulted in at least 2-fold increases in median blood and exhaled breath TTHM across two study groups, regardless of whether the study site was characterized by high (NC site median = 136 μg/L) or low (TX site median = 38 μg/L) TTHM in the residential water supply. This magnitude of increase was observed for all THM species except CHBr₃ in the blood samples, but only for CHCl₃ in the exhaled breath samples. We also observed greater than 2-fold increases in median exhaled breath concentrations of CHCl₃ at both sites, and in blood CHCl₃ and TTHM in two of the three participants at the TX site for the hand dishwashing activities. There was no activity without a 2-fold increase in concentration in any biomarker of exposure for at least one THM and one individual.

The greatest observed increase in blood and exhaled breath THM concentration in any participant was due to showering and bathing. The average increases in blood TTHM due to showering were 358 and 79 pg/mL at the NC and TX sites, respectively. Average increases due to bathing were 164 and 118 pg/mL, and due to hand dishwashing were 98 and 57 pg/mL, respectively. However, we observed a high degree of inter-participant variation in the increase due to hand dishwashing at both sites. Increases in blood TTHM concentration due to other activities were less than 20 pg/mL, and were also highly variable. More human-based research is needed to determine whether acute and frequent exposures to THM at these concentrations have public health implications.

The results of our study are consistent with findings of other studies for which shower water and pre- and post-shower blood THM concentrations have been reported. Table 5 presents a summary of shower water and participant blood (pre- and post-shower) THM concentrations for two studies in addition to ours. If we group the shower water concentrations of CHBrCl₂ for the 5 study sites described in Table 5 into 3 categories: 6, 11-14, and 33 µg/L, the corresponding median blood CHBrCl₂ concentrations reported for these groups are 19, 28-43, and 93 pg/mL after showering for 10 minutes. These findings indicate a dose-response between concentration in the source water and blood. Similar correspondence between shower water and post-shower blood CHBr₂Cl and CHCl₃ concentrations were observed across the five study sites, as well as for source water and post-bathing THM concentrations reported for our study and the study by Backer et al. (data not shown). Lynberg et al. (2001) did not conduct a bathing analysis.

Our observations are also consistent with results reported in other residential studies of exposures to disinfected tap water in which air and exhaled breath samples were analyzed for THM. Table 6 is a summary of results of during-shower air THM concentrations from three studies (Egorov et al., 2003; Kerger et al., 2000; May et al., 1995) in addition to ours. THM concentrations of exhaled breath from participants during showering were also reported by Egorov et al. (2003). In all cases reported in Table 6, the air concentrations during showers showed the same decreasing trend of $CHCl_2 > CHBrCl_2 > CHBr_2Cl$, which was consistent with their relative concentrations in the source water of each respective study. When we adjust for variation in THM water concentrations across the studies by taking the ratios of the shower air to source water concentrations, this ratio is roughly 2.2 and 2.4 μ g/m³ per μ g/L water for the "high" and "low" sites in our study compared to a ratio of 1.7 μ g/m³ per μ g/L water obtained from the May et al. (1995) and Egorov et al. (2003) data, and a value of 3.5 μ g/m³ per μ g/L water

obtained from the Kerger et al.(2000) data. The differences in ratios between these studies could be due to a variety of factors known to affect THM transfer coefficients from water to air that were not taken into account in this comparison. These factors include water temperature and flow rate, shower duration, volume of shower enclosure, air exchange rates, and showerhead type. Available published studies on the measurement of THM concentrations in exhaled breath are sparse. Table 6 summarizes our results for the "high" and "low" sites along with values presented by Egorov et al. (2003) from their recent study of exposures to tap water DBPs in a Russian city. In each case, the data for the three THM listed show a corresponding gradient, high to low, between the during-shower air concentrations and the post-shower exhaled breath concentrations. However, both our "high" site and "low" site concentrations for breath CHCl₃ are significantly lower than the value reported by Egorov et al. (2003) despite the relatively close agreement between air concentrations at our "high" site and their value (cf. Table 6). A reason for the observed differences could be the time when the samples were taken after exposure ended (Gordon et al. 1998; Weisel et al., 1999; Xu and Weisel, 2005). In our study, breath samples were taken 5 minutes after exposure ceased; in the Egorov et al. study, breath samples were collected within one minute after subjects completed their showering activity.

We observed changes in baseline (cold tap water) THM concentrations over the course of each study day. However, the difference between baseline concentration for any THM by time of day was not statistically significant across the study population (p = 0.07 to 0.65). We also observed a high degree of variation between tap water THM concentrations over the period of study, especially at the NC site. For example, at this site water samples were collected 7 different days over the period of approximately 43 days, and the range in TTHM concentrations in the samples collected at 8:00 a.m. on each of those days was 139 to 200 μ g/L (average = 169), and the

maximum CHBrCl₂ was 63 μ g/L (range 23 to 63). The THM levels in our samples were much different than the average concentrations reported by the utility that provides water to our NC study site. For example, the utility reported an annual average TTHM concentration of 76.7 μ g/L (range, 28 to 145), and a maximum CHBrCl₂ concentration of 17 μ g/L (range, 5 to 17) for the year in which our study was conducted. These finding are important in terms of exposure assessment for epidemiological studies concerning THM, because they indicate that while "snapshot" measurements of THM on a given day can be representative of levels for water use activities on that day, they may not be representative of THM in a specific residential water supply over a longer period of time.

The results of our study support the findings of other studies that blood THM concentrations in response to equal or equivalent THM exposure appear to be higher in some individuals. At each of our study sites, we observed a large difference in relative increase in THM blood levels by one of the study participants in response to exposure by showering in waters with approximately the same THM concentration and temperature. We also observed differences in response for the same individual to exposure from hand dishwashing. Although our sample size is very small, these findings lend support to similar patterns reported by Backer et al (2000) and Lynberg et al. (2001). Backer et al. suggested that such differentiation in response may be the result of differences in individuals' abilities to metabolize THM. A number of metabolic enzymes exist in polymorphic form. For example, some THM are substrates for glutathione S-transferase -theta-1 (GSTT1)-mediated glutathione conjugation reactions (Landi et al. 1999). Among Caucasian populations, about 17% to 18% of people are null for this gene. Another candidate enzyme is CYP2E1, which has a demonstrated role in THM metabolism of THM (Allis et al. 2001; Constan

et al. 1999). Further research is needed to understand the implication of these findings in terms of design of epidemiology studies concerning trihalomethanes.

The findings of our study have important ramifications for exposure assessment in epidemiologic studies concerning trihalomethanes. The study confirms that showering and bathing activities are important sources of THM exposure. It provides evidence that hand dishwashing, indirect shower exposure, and other hot water use activities could also be important sources, but need more study. Water temperature and THM concentration, and duration of use have been demonstrated to be important variables for quantifying THM exposure during showering and bathing (Giardino and Andelman 1996; Keating et al., 1997; Kerger et al., 2000; Wilkes et al., 2004). Water temperature was not correlated to water THM concentration in our study. It is well established that THM concentrations of water in residential water heaters are generally much higher than in tap water from the utility distribution system, and we observed much higher temperatures in activity-associated water compared to baseline (cold tap) samples. However, we observed THM concentration ratios (total and all species) near or below 1.0 between these water samples for most all activities. THM concentrations in air samples collected in association with these water use activities were all significantly elevated, indicating that THM formed by heating of the water supply were volatile. For example, showering and indirect shower exposure median air concentrations were 318 and 142 g/m³ compared to a baseline of 4 and 3 g/m³, respectively at our NC site (cf. Table 2). The fact that the ratios of the shower air to source water concentrations for the "high" and "low" sites were about equal (2.2 and 2.4) in our study indicates that estimates of air THM concentrations associated with specific hot water use activities may be possible if accurate THM water concentrations are known. Weisel and Chen (1994) observed a doubling of ChCl₃ concentration, and a 50% increase in CHBrCl₂ and

CHBr₂Cl in water heated to 65 C that contained 0.7 to 0.8 mg/L total chlorine residual. They reported that a majority of this increase occurred within a half hour, and was essential complete within one hour. If THM concentrations do "plateau" in a residential hot water heater, obtaining measurements of temperature and THM concentration in separate hot and cold water samples during an epidemiology study could simplify exposure assessment. The temperature measurements could be used to estimate potential range of dermal exposure. Gordon et al. (1998) reported a strong effect of bathwater temperature on dermal absorption of chloroform, and it is likely this effect would hold for other hot water uses with dermal contact. Likewise, it might be possible to estimate air THM concentrations for specific water use activities based on the hot and cold water THM concentration. These results could be used in conjunction with air to water THM concentration ratios to construct "confidence intervals" for predictions of air THM concentrations from specific water use activities. A limitation to this approach is that these ratios can vary by activity as a function of room volume, ventilation, and other factors. For example, in our study inter-site differences in these factors were minimized for the shower activity, and the ratios were near-equal (2.2 and 2.4). However, the average air to water ChCl₃ concentration for the bathing activities, which were measured in the bathroom rather than shower stall, were 0.7 at our NC site and 1.2 at the TX site. The inter-site difference in ratios for the bathing activity was due to a difference in bathroom volume. More research is needed to determine if standardized air to water THM concentration ratios for hot water activities related to significant THM exposure can be developed, and applied in the context of an epidemiologic study.

The results of our study clearly indicate that epidemiology studies concerning trihalomethanes need to consider hot water use activities as important exposure events. Further research is needed

in designing epidemiologic studies that minimize data collection burden, yet maximize accuracy in classification of dermal and inhalation THM exposure during these activities.

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Table 1. Description of water use activities and duration over the course of the study.

DAY 1		DAY 2 (1 week after Day 1)									
Time	Water Use Activity	Duration (minutes)	Time	Water Use Activity	Duration (minutes)						
21:00 ^a	Participant arrives at the study house, and sleeps there overnight		21:00 ^a	Participant arrives at the study house, and sleeps there overnight							
8:00	Baseline measurements: ambient household air, tap water, blood THM	6.0	8:00	Baseline measurements: ambient household air, tap water, blood THM	6.0						
8:20	Breakfast, including preparation and consumption of a hot beverage from tap water (0.25 liters)	25.0	8:20	Breakfast, including consumption of a cold beverage prepared from tap water (0.25 liters)	25.0						
10:00	Hot water shower ^b	13.0 °	10:00	Hot water bath ^b	23.0 ^g						
13:00	Lunch, including drinking 0.5 liters of cold tap water	30.0	13:00	Lunch, including consumption of bottled water (no specified volume) ^f	er 30.0						
15:00	Automatic clothes washing (Clothes Washer) ^d	50.0	14:00	Automatic clothes washing, adding bleach du the wash cycle (Clothes Washer II) ^d	ring 50.0						
17:30	Hand washing ^e	0.5	16:00	Hand washing of dishes h	10.0						
18:00	Supper, including consumption of bottled water (no specified volume) ^f	45.0	18:00	Supper, including consumption of bottled water (no specified volume) ^f	45.0						
19:00	Automatic dish washing, open dishwasher at end of cycle	50.0	19:00	Sitting in room adjacent to the study bathroom and a shower event, opening bathroom door a							
21:00	Participant departs study house		21:00	Participant departs study house							

^a evening before day of study; arrival between 21:00 and 23:00 allowed
^b No cleaning products such as soap or shampoo used by the participant; subjects wore swimsuits
^c Participant in shower stall or bath for 10 minutes, followed by 3 minutes in study bathroom with door closed for changing clothes

d Participant did not stay in same room as water use device e No cleaning products such as soap used by the participant

^f Bottle water was tested and confirmed that no THM species were present.

g Filling time from 10:00 to 10:06, maintained constant (6 minutes) for each participant; this was sufficient volume to submerge the torso and legs; participant stayed in the tub from 10:06 to 10:20 (14 minutes); followed by 3 minutes in study bathroom with door closed for changing clothes; subjects were swimsuits

^h Detergent (Dawn Ultra) was used.

ⁱ Termed "indirect shower exposure"

Table 2: Median temperature and concentration of THM in air $(\mu g/m^3)$ for baseline and activities with at least a 2-fold increase in blood concentration for at least one participant.

•	•	Air Temp °C		CHCl ₃		CHBrCl ₂		CHBr ₂ Cl		$CHBr_3$		TTHM	
		NC Site	TX Site	NC Site	TX Site	NC Site	TX Site	NC Site	TX Site	NC Site	TX Site	NC Site	TX Site
B	aseline Day 1	24	24	4	2	3	2	BDL^a	BDL^a	BDL^{a}	BDL^a	8.0	5.0
1	Range	22-24	23-25	2-10	1-2	BDL-7	2-3	_b	_b	_b	_b	5-19	5-7
H	ot Beverage	24	23	7	2	2	3	BDL	BDL	BDL	BDL	10	6
F	Range	24-25	23-24	3-10	2-2	1-4	2-3	-	-	-	=	6-16	6-7
Sł	nower	25	24	318	67	54	23	9	4	BDL	BDL	384	95
F	Range	24-32	20-28	219-351	50-70	31-68	20-25	4-13	3-6	-	=	255-431	74-102
\mathbf{C}	othes Washer	24	27	21	4	7	2	BDL	BDL	BDL	BDL	31	4
F	Range	24-27	25-27	7-25	2-5	BDL-8	0.7-3	BDL-2	-	-	-	9-34	2-5
H	and Washing	24	23	49	3	10	2	2	BDL	BDL	BDL	62	6
F	Range	22-27	22-23	19-85	3-5	3-13	1.3-2.3	BDL-2	-	-	-	23-101	6-9
A	uto Dishwashing	24	25	8	5	2	3	BDL	BDL	BDL	BDL	11	9
I	Range	24-25	24-26	4-12	4-5	BDL-3	3-3	-	-	-	-	6-18	9-10
Ва	aseline Day 2	24	23	3	1.0	0.8	1.3	BDL	BDL	BDL	BDL	6	4
F	Range	23-24	21-24	2-4	0.8-2	BDL-1	1-3	-	-	-	-	4-7	4-7
Ва	ath	24	23	71	14	12	7	2	1.4	BDL	BDL	88	24
F	Range	22-24	21-24	49-98	8-61	9-14	4-15	1-3	BDL-2	-	-	60-112	13-79
\mathbf{C}	othes Washer II	24	27	9	9	2	2	BDL	BDL	BDL	BDL	12	14
F	Range	24-25	27-28	8-33	4-13	1-5	0.9-3	-	-	-	=	11-39	6-17
H	and Dishwashing	24	24	8	5	2	1	BDL	BDL	BDL	BDL	11	8
F	Range	24-25	24-28	6-17	3-9	1-4	1-5	-	-	-	=	9-23	6-15
In	direct Shower Exp	24	24	142	75	30	27	7	5	BDL	BDL	176	108
I	Range	22-25	22-24	117-370	63-86	20-114	25-29	3-11	3-7	-	-	151-495	100-115
				_	_		_		_				

^a BDL = below detection limit. Detection limits are 0.5 μ g/m³ for CHCl₃, 0.7 μ g/m³ for CHBrCl₂, 0.8 μ g/m³ for CHBr₂Cl, and 1.0 μ g/m³ for CHBr₃. ^b Ranges not included as all samples were at or below detection.

 $Table\ 3.\ Median\ THM\ concentration\ in\ blood\ (pg/mL)\ approximately\ 5\ minutes\ before\ and\ after\ water\ use\ activities.$

CHCl₃ NC Site		TX Si	CHBrCl ₂ Site NC Site		CHBr ₂ Cl TX Site NC Site TX		TX Sit	CHBR ₃ TX Site NC Site			TTHM TX Site NC Sit									
Activity Hot Bev. Range	PRE 40 34-44	31	PRE 19 8-22	POST 13 9-16	PRE 9 6-17	POST 8 5-15	PRE 4 4-8	POST 3 3-9	PRE 2 1-5	POST 2 0.8-5	PRE 2 1-4	POST 1 1-4	PRE 0.6 0.5-1	POST 0.6 0.5-1		POST 0.6	PRE 52 41-64	POST 44 36-52		POST 21 13-26
Shower Range	26 23-83	290 262-374	13 11-13	63 56-66	6 3-8	93 64-95	4 3-7	28 26-31	1 0.6-3	13 12-18	1 0.9-3	6 6-10	0.7 0.5-1	0.8 0.5-1	0.5 0.5-0.6	0.7 0.6-1	34 31-90	399 338-482	18 16-23	97 88-108
Lunch w/ water	51	45	37	41	11	12	6	7	2	3	2	2	0.6	0.6	0.6	0.6	66	59	45	48
Range	38-99	43-54	18-44	33-41	9-14	9-13	5-12	5-9	2-3	2-3	1-5	1-4	0.5-1	0.5-1	0.5-0.8	0.6-0.7	51-110	57-70	25-62	47-51
Clothes washer I	32	52	27	35	7	12	5	5	2	2	2	2	0.6	0.5	0.6	0.5	43	67	35	42
Range	30-44	51-166	19-43	19-45	5-9	8-14	4-9	2-8	1-2	1-3	1-4	0.8-4	0.5-0.9	0.5-0.8	0.5-0.6	0.5-0.7	39-50	66-175	25-56	22-58
Hand wash Range	h 36 27-48	48 34-51	23 17-33	19 11-43	9 5-10	11 6-13	4 3-8	5 3-8	2 0.8-2	2 0.9-3	1 0.9-3	1 0.8-3	0.5 0.5-0.6	0.6 0.5-0.6	0.6 0.5-0.7	0.6 0.5-0.6	47 33-61	61 41-65	29 21-39	25 15-31
Auto Dish-wash	32	38	17	29	8	9	4	4	2	2	1	1	0.6	0.5	0.6	0.5	42	49	21	40
Range	22-36	30-43	14-43	17-39	4-9	6-11	3-4	4-4	0.7-2	0.8-3	0.9-5	1-3	0.6-0.6	0.5-0.6	0.5-1	0.5-0.5	27-47	37-56	20-62	22-45
Cold Bev. Range	30 24-95	40 29-56	21 20-50	24 16-85	7 3-47	6 5-24	5 4-8	4 3-9	2 0.5-17	2 0.8-9	1 1.0-3	1 0.6-3	0.6 0.5-0.9	0.5 0.5-0.8	0.5 0.5-0.6	0.5 0.5-0.5	39 29-161	48 37-88	27 26-62	36 21-89
Bath Range	37 27-40	161 125-188	12 8 8-22	54 48-156	5 5-14	41 40-43	3 2-7	36 26-65	1 1-5	10 6-13	1 0.5-3	10 8-11	0.6 0.5-0.9	0.7 0.5-1	0.5 0.5-0.5	1 0.5-1	44 35-60	212 181-234	16 12-32	101 83-231
Clothes washer II	33	52	22	17	5	8	8	5	2	2	2	2	0.5	0.6	0.5	0.5	44	66	34	17
Range	22-44	38-61	12-39	-	5-12	8-14	4-8	5-8	0.8-3	1-4	0.9-3	1-2	0.5-0.8	0.5-1	0.5-0.5	0.5-0.6	30-50	50-72	18-50	7-24
Hand dishwash	43	73	33	42	7	19	4	12	2	6	1	3	0.6	0.6	0.6	0.7	56	99	38	58
Range	39-48	41-285	9-41	25-97	5-15	8-63	3-9	7-66	0.7-4	2-11	0.5-3	1.1-18.1	0.5-1	0.5-0.7	0.5-0.6	0.5-2	45-60	52-359	13-53	33-183
Indirect Shower Ex	35	50	52	19	6	10	5	6	1	2	1	2	0.6	0.5	0.5	0.5	45	63	53	23
Range	28-43	45-59	15-52	12-61	5-11	6-15	3-9	3-9	1-4	0.8-4	0.6-3	0.6-3	0.5-0.6	0.5-1	0.5-0.5	0.5-0.6	36-53	53-70	21-57	19-73

Table 4. Median and range of THM concentrations (ug/m³) in exhaled air: baseline and post-water activity by study site.

	CHCl ₃		CHBrCl ₂		CHBr ₂ Cl ^a	I	CHBr ₃ ^a	- J J	TTHM	TTHM		
	NC Site	TX Site	NC Site	TX Site	NC Site	TX Site	NC Site	TX Site	NC Site	TX Site		
Baseline Day 1	5	1	2	2	BDL^{a}	BDL^a	BDL^a	BDL^{a}	9	6		
Range	2-6	1-2	BDL-5	2-3	_b	_b	_b	_b	4-13	5-6		
Hot Beverage	4	2	2	3	BDL	BDL	BDL	BDL	7	7		
Range	2-5	0.8-5	BDL-5	1-4	-	-	-	-	5-14	6-8		
Shower	24	6	6	3	BDL	BDL	BDL	BDL	28	11		
Range	16-51	5-8	2-8	3-4	-	-	-	-	26-61	9-14		
Clothes Washer	11	1	3	1	BDL	BDL	BDL	BDL	15	4		
Range	3-17	0.7-2	BDL-6	BDL-2	-	-	-	-	6-25	4-5		
Hand Washing	6	1	2	2	BDL	BDL	BDL	BDL	9	5		
Range	3-11	0.9-1	BDL-2	1-5	-	-	-	-	5-15	4-12		
Auto Dishwashing	4	3	1	2	BDL	BDL	BDL	BDL	7	5		
Range	2-4	3-4	BDL-2	2-2	-	-	_	-	5-15	4-12		
Baseline Day 2	5	1	2	0.7	BDL	BDL	BDL	BDL	9	4		
Range	2-12	BDL-2	1-9	BDL-2	-	-	-	-	6-15	3-6		
Bath	15	7	3	3	BDL	BDL	BDL	BDL	20	13		
Range	11-22	4-9	1-4	3-3	-	-	-	-	14-26	9-13		
Clothes Washer II	12	2	2	2	BDL	BDL	BDL	BDL	16	6		
Range	6-13	2-3.5	1-8	1-2	-	-	-	-	9-46	5-7		
Hand Dishwashing	14	3	2	2	BDL	BDL	BDL	BDL	18	7		
Range	5-18	3-4	BDL-3	1-5	-	-	-	-	7-22	6-11		
Indirect Shower Exp	5	2	0.8	2	BDL	BDL	BDL	BDL	8	6		
Range	2-8	1-2	BDL-2	2-2	-	-	-	-	4-11	5-6		

^a BDL = below detection limit. Detection limits are 0.5 μ g/m³ for CHCl₃, 0.7 μ g/m³ for CHBrCl₂, 0.8 μ g/m³ for CHBr₂Cl, and 1.0 μ g/m³ for CHBr₃. ^b Ranges not included as all samples were at or below detection.

Table 5. Comparison of median shower water, pre- and post-shower blood THM concentrations for participants in three studies

	Sh	ower W	ater Conc (ug/L)	entration	1	Post-s		Blood Co g/mL) ^a	ncentrat	Ratio: Post- to Pre-Shower Blood Concentration					
THM	Backer	Lynbe	erg et al.			Backer	Lynbe	erg et al.			Backer	Lynbe	erg et al.		
compoundb	et al.	(20	$(001)^{c}$	Our st	tudy ^c	et al.	(20	$(001)^{c}$	Our st	tudy ^c	et al.	(20	$(001)^{c}$	Our	study ^c
	$(2000)^{c}$					$(2000)^{c}$					$(2000)^{c}$				
		HIGH	LOW	HIGH	LOW		HIGH	LOW	HIGH	LOW		HIGH	LOW	HIGH	LOW
		SITE	SITE	SITE	SITE		SITE	SITE	SITE	SITE		SITE	SITE	SITE	SITE
CHCl ₃	28	85	8	148	28	120	280	57	290	63	4	3	7	2	2
CHBrCl ₂	6	14	12	33	11	21	38	43	93	28	4	3	4	3	3
CHBr ₂ Cl	1	14	2	6	2	5	41	6	13	6	5	3	3	2	3

a approximately 10 minutes post-shower
 b bromoform was above below or near detection limit in water source at 4 of 5 sites, and thus not comparable
 N = 11 in Backer et al. study; N = 25 at each site of Lynberg et al. study; N = 4 and 3 at our HIGH and LOW site, respectively

Table 6. Comparison of THM concentrations in source water, during-shower air, and post-shower breath concentrations in this and other published studies.^a

	Source Water Concentration (µg/L)						g-Shower	Air Conce	entratio	$(\mu g/m^3)^b$	Post-S	Post-Shower Breath Concentration (µg/m³) ^c					
THM Compound ^d	May	Kerger	Egorov	This S	Study ^e	May	Kerger	Egorov	This	Study ^e	May	Kerger	Egorov	This	Study ^e		
	et al.	et al.	et al.	High	Low	et al.	et al.	et al.	High	Low	et al.	et al.	et al.	High	Low		
	$(1995)^{e,f}$	$(2000)^{e}$	$(2003)^{e}$	Site	Site	1995) ^{e,f}	$(2000)^{e}$	$(2003)^{e}$	Site	Site	$(1995)^{e}$	$(2000)^{e}$	$(2003)^{e}$	Site	Site		
CHCl ₃	51	47	198	148	28	84	165	330	318	67	_	_	110	24	6		
CHBrCl ₂	17	42	7	33	11	24	80	8	54	23	_	_	1	6	3		
CHBr ₂ Cl	6	31	1	6	2	nd	16	nd	9	4	_	_	nd	1	1		

^a Kerger et al. and Egorov et al. reported mean concentrations; May et al. reported median concentrations; and we report median concentrations from Tables 2, 3, and 5.

^b Shower duration: May et al. reported 10 min; Kerger et al. reported 6.8 min and 12 min; Egorov et al. reported 15-20 min; we report 10 min.

^c Breath sample collection: Egorov et al. reported 1 min post-exposure; we report 5 min post-exposure.

^d In water source, CHBr₃ was near or below limit of detection at most sites; in air samples, CHBr₂Cl and CHBr₃ were below limits of detection in Egorov et al. and May et al. studies; in breath samples, CHBr₂Cl and CHBr₃ were below limits of detection in Egorov et al. and this study.

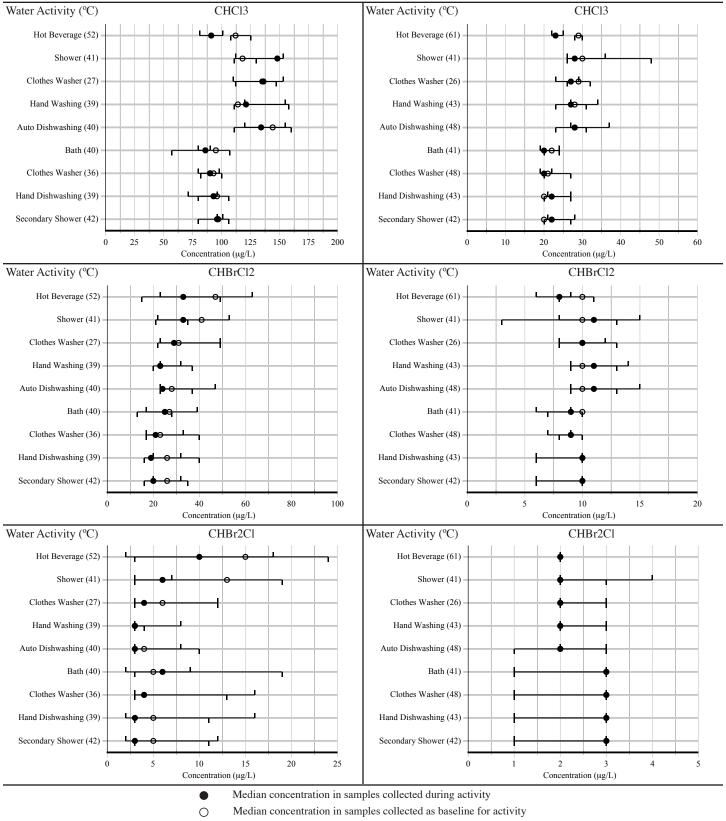
^e N = 44 in May et al. study; N = 20 for source water and N = 12 for shower air in Kerger et al. study; N = 14 for source water, N = 35 for shower air, and N = 9 for exhaled breath in Egorov et al. study; N = 4 and 3 for source water, shower air, and exhaled breath at our "High" and "Low" site, respectively.

f Median values for CHCl₃, CHBrCl₂, and CHBr₂Cl for source water and shower air estimated from plots in May et al.

Figure 1. Median and range of THM concentrations in tap water (:g/L)

Notes:

- 1. If only occurs on a graph, the median concentration in samples collected during the activity was approximately equal to that in the samples collected as baseline for the activity.
- 2. All concentrations rounded to nearest integer for presentation purposes.
- 3. Concentration scales used vary by study site and THM compound.



Median concentration in samples collected during activity
Median concentration in samples collected as baseline for activity
Range in concentration of samples collected during activity
Range in concentration of samples collected as baseline for activity

(°C) Median ambient water temperature during activity